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Indoor Air Exposure to Coal and Wood Combustion Emissions Associated with a High Lung Cancer Rate in Xuan Wei, China

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16. ABSTRACT
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Residents of Xuan Wei County in China have unusually high lung cancer mortality that cannot be attributed to tobacco use or occupational exposure. They are exposed to smoke from unvented, open pit coal or wood fires (often used for cooking and heating). The variation in lung cancer rates among communes within the county suggests that indoor combustion of smoky coal may be the prime determinant of lung cancer. To characterize the air in Xuan Wei homes, samples of air particles and semivolatile organic compounds were collected from homes located in two communes; one commune has a high rate of lung cancer, and the other has a low rate. Samples collected in the commune where the lung cancer rate is high and where smoky coal is the predominant fuel contained high concentrations of small particles with high organic content; organic extracts of these samples were mutagenic. Samples from homes in the wood-burning commune, which has a low rate of lung cancer, consisted mostly of larger particles of lower organic content and mutagenicity. The smoky coal sample was a mouse skin carcinogen and was a more potent initiator of skin tumors in comparison to the wood or smokeless coal sample.

EPIDEMIOLOGIC BACKGROUND

Xuan Wei is a rural county in Yunnan Province, China, with a population of one million. More than 40% of the males, but less than 0.1% of females, smoke tobacco. Although there are some local industries, including coal mining, electric power generation, and light manufacturing, more than 90% of the local residents are farmers. Xuan Wei residents traditionally have used one of three fuel types — "smoky" coal, "smokeless" coal, or wood — for domestic heating and cooking. Smoky coal is comparable to medium-volatile, low-sulfur (0.2%) bituminous coal with a heating value of 27.1 MJ/kg and 20% ash. Smokeless coal is a low-grade (49% ash), high-sulfur coal (1.9%) with a heating value of 14.5 MJ/kg. Until recently it was customary to burn domestic fuel in a shallow, unvented pit in the floor of the family dwelling's main room. This custom, which still persists in some parts of the county, is the reason for the very high levels of indoor air pollution. Domestic smoky coal and smokeless coal are generally obtained from small mines, scattered throughout the county, as shown in Fig. 1.

In this report, the term "lung cancer" includes carcinomas of lung parenchyma, bronchus, and trachea. The annual lung cancer mortality rates are shown in Table 1. Female lung cancer mortality in Xuan Wei was China's highest, and that of males was among China's highest (Office of Tumor Preventive Research 1979). In Xuan Wei, death caused by lung cancer was the only cancer mortality statistic that exceeded the Chinese national average

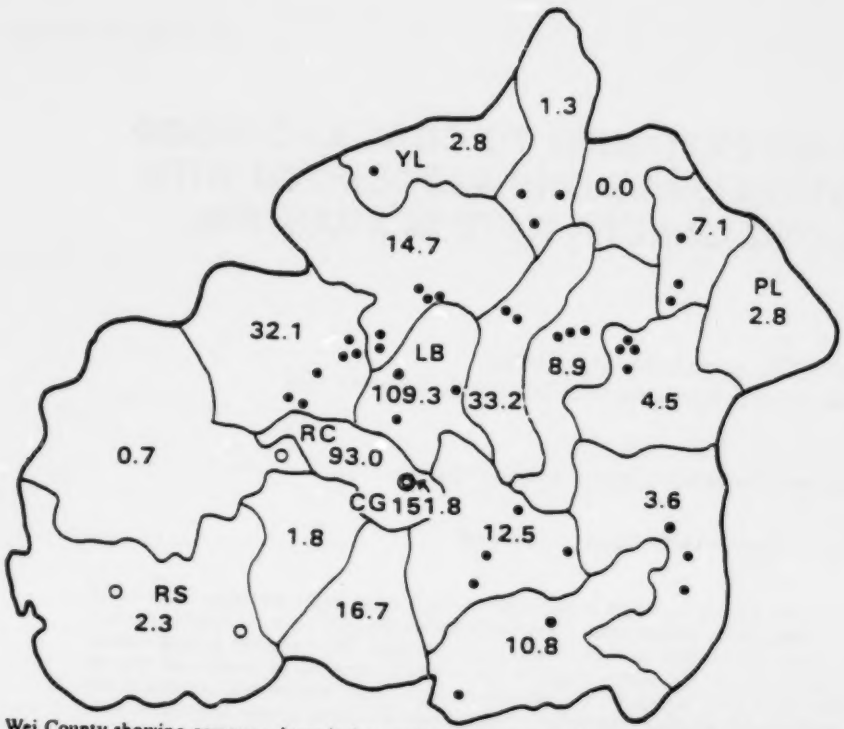

Fig. 1. Map of Xuan Wei County showing commune boundaries, each commune's unadjusted annual lung cancer mortality rate per 100,000 (both sexes, 1973-1975) and mines supplying domestic coal: (•) = smoky coal, (o) = smokeless coal. Designated communes with high mortality: CG (Cheng Guan), LB (Lai Bin), and RC (Rong Cheng); those with low mortality: PL (Pu Li), RS (Re Shui), and YL (Yang Liu). Taken from Mumford et al. (1987).

(Editorial Committee for the Atlas of Cancer Mortality in the People's Republic of China 1979). A clinical survey conducted in Xuan Wei in the 1970s showed that of 115 tumor specimens examined pathologically, 58% were squamous cell carcinomas, 28% were adenocarcinomas, and the remaining 14% were mixed-cell, undifferentiated, or alveolar carcinomas. The case detection rate was higher in farmers than in coal miners or office workers, suggesting that lung cancer in Xuan Wei is not associated with nonagricultural occupations.

As shown in Fig. 1 (Mumford et al. 1987), there is wide variation across Xuan Wei communes in unadjusted annual lung cancer mortality rates. Rates are highest in the central communes of Cheng Guan (CG), Lai Bin (LB), and Rong Cheng (RC). In these communes, smoky coal is used in most homes. From 1973 to 1975, the average mortality rates in communes with smoky coal mines was 34.7 per 100 000, as compared to 4.1 in communes without smoky coal mines. As shown in Fig. 2, lung cancer mortality in each of 11 communes from 1973 to 1975 was highly correlated with the corresponding percentage

Table 1. Annual lung mortality rates in China and the United States.

	Mortality rate (per 100,000)						
	Unadjusted			Age-adjusted to 1964 China population		Age-adjusted to 1970 U.S. population	
	Males	Females	Combined	Males	Females	Males	Females
China (1973-75)			5.0	6.8	3.2	12.3	5.7
United States (1970)	53 7	12.0		30.0	6.3	53.7	12.0
Yunnan Province (1973-75)			2.8	4.3	1.5	6.9	2.5
Xuan Wei County (1973-79)	27.0	24.5		27.7	25.3	43.2	38.7
Three high-mortality Xuan Wei communes, Cheng Guan, Lai Bin, Rong Cheng (1973-79)	114.4	120.6		118.0	125.6	186.8	193.4

* Taken from Mumford et al. 1987.

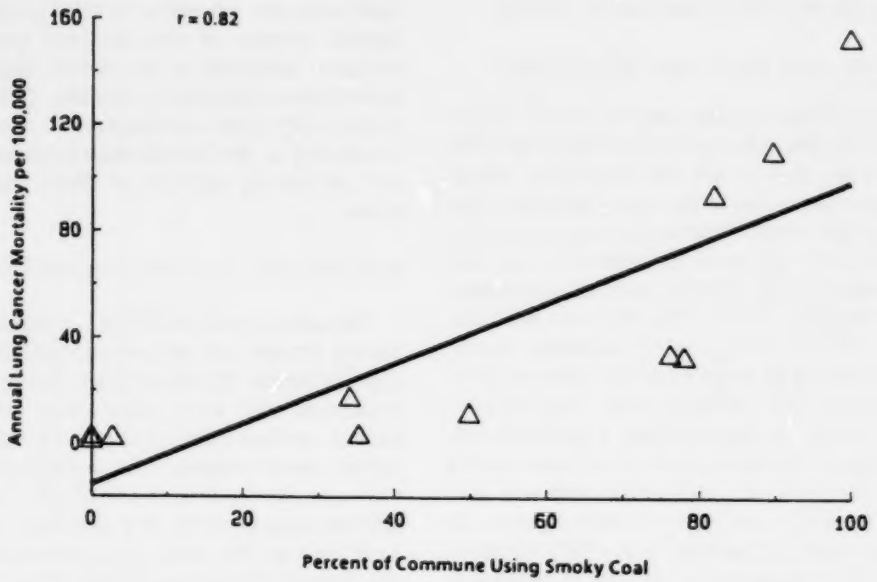

Fig. 2. Percentage of households burning smoky coal before 1958 and unadjusted annual lung cancer mortality in 11 Xuan Wei communes during 1973-75.

of households in which smoky coal was used before 1958 (r=0.82). Tobacco smoking habits, fuel use distributions, and lung cancer mortalities in LB and Re Shui (RS) communes are presented in Table 2. Although the smoking rate was higher in RS, smoky coal use and lung cancer mortality were higher in LB.

These observations have generated the hypothesis that indoor air pollution from smoky coal is the prime determinant of lung cancer in Xuan Wei, particularly in women, who generally start the fire and cook. Xuan Wei, with its wide variation in mortality rates, its high indoor pollution exposures, and its stable, rural population, presents an unusual opportunity to test this hypothesis. Chinese and American investigators have mounted interdisciplinary studies to relate Xuan Wei's indoor air pollution to lung cancer.

This paper reports the characterization of air in Xuan Wei homes during coal and wood combustion, the linkage of these results to the epidemiological

Table 2. Tobacco smoking and fuel use in Lai Bin and Re Shui communes.

	Commune	
	Lai Bin	Re Shui
Approximate population	60,000	60,000
Percent of population ever smoking tobacco		
Males	44.0%	49.1%
Females	0.05	0.07
Percent of population using:		
Smoky coal	89.7%	0.0%
Smokeless coal	1.6	33.4
Wood	8.7	66.6
Unadjusted annual lung cancer mortality per 100,000 (1973-75)	109.3	2.3

data, and an evaluation of how well the short-term bioassays can predict the cancer risk in humans.

AIR SAMPLING, ANALYSES, AND BIOASSAYS

Indoor and outdoor air was sampled in CG, where mortality is Xuan Wei's highest and smoky coal is the predominant fuel, and in the RS commune, where mortality is low and wood (67%) and smokeless coal (33%) are the fuels used. Four homes burning smoky coal and wood were selected from both CG and RS communes, respectively. Smokeless coal was burned in one home selected in CG. The emission particles smaller than 10 μm (PM_{10}) were collected during three daily cooking periods by high-volume (HV) samplers (sampling at 1.2 m^3/min) with 10-μm size-selective inlets (SSIs). At one CG home, a medium-volume (MV) sampler (sampling at 0.11 m^3/min) with a PM_{10} SSI, a filter, and an XAD-2 resin trap was used to collect both particles and semivolatile organics, as described previously (Mumford et al. 1987). Outdoor HV sampling was conducted in one site in each commune. Carbon monoxide was also measured by using SPE CO detectors (General Electric Company 1980) in three selected homes that burned different fuel types. The indoor air particles were examined for size and physical properties (by scanning electron microscopy), the percentage of extractable organic mass (by dichloromethane extraction), and the polynuclear aromatic hydrocarbon (PAH) content (by GC/MS; Nishioka et al. 1982). The organic extracts were also fractionated into compound classes to identify the mutagenic components (Petersen and Chuang 1982). The Ames Salmonella mutation assay (Ames et al. 1975; Claxton 1981) was employed to determine the mutagenicity of the extracts and fractions. For carcinogenicity, skin tumor initiation/promotion studies were conducted in Sencar mice (40 female mice per treated or control group) by using the organic extract of the coal and wood combustion emission particles as the tumor initiator and 12-*O*-tetradecanoylphorbol-13-acetate (TPA) as the promotor. Complete carcinogenesis studies were also conducted in the Sencar mice to assess the initiating and promoting activity of these combustion emissions.

PARTICULATE AND CO CONCENTRATIONS

The concentrations of PM_{10} particles inside homes during smoky coal and wood combustion were very high, whereas concentrations during combustion of smokeless coal were much lower (Table 3). As expected, outdoor concentrations were much lower than indoor concentrations. The outdoor concentrations in CG were higher than those in RS. Particle concentrations measured by HV and MV samplers agreed well, except for those concentrations measured in RS. Indoor CO concentrations were high, especially during the combustion of smoky coal. CO concentrations were 10 to 220 μL/L, 20 to 150 μL/L, and 40 to 95 μL/L during combustion of smoky coal, smokeless coal, and wood, respectively.

PHYSICAL AND CHEMICAL CHARACTERIZATION

Of the particles from smoky coal combustion, 51% were less than 1 μm in diameter; 49% were between 1 and 10 μm. Most particles were spherical. In contrast, 94% of the particles from wood combustion ranged from 1 to 30 μm in diameter; only 6% were less than 1 μm. The emission particles from smoky coal combustion contained high concentrations of organic matter, whereas the samples from the other

Table 3. Particle concentrations (PM_{10}) and organic extractable mass in Xuan Wei homes during cooking*.

Fuel	Particle concentration (mg/m3)			Extractable organic mass (%) of particles		
	Indoor		Outdoor	Indoor		Outdoor
	HV	MV	HV	HV	MV	HV
Smoky coal	24.4 ± 3.3	25.8	0.58	72.3 ± 6.7	76.2	42.0
Wood	22.3 ± 2.0	10.9	0.05	55.1 ± 7.0	63.1	13.3
Smokeless coal	1.8	1.1	-	27.0	33.3	-
Laboratory (in CG)	-	-	0.32	-	-	14.1

* Results from high-volume (HV) and medium-volume (MV) particulate sampling. Values are mean ± SEM (where shown).

Table 5. Organic mass distribution and mutagenicity of indoor air particles (PM_{10}) during cooking in Xuan Wei homes.

Fraction	Smoky Coal		Wood		Smokeless Coal	
	Mass (%)	Mutagenicity[b] ($\times 10^3$rev./m^3)	Mass (%)	Mutagenicity ($\times 10^3$rev./m^3)	Mass (%)	Mutagenicity ($\times 10^3$rev./m^3)
Neat sample	100	58.9	100	11.1	100	1.3
Aliphatics (hexane)	5.0	0	1.5	0	3.2	0
Aromatics (hexane/benzene)	36.4	18.1	7.7	3.1	29.7	0.5
Moderately polar compounds (dichloromethane)	35.0	8.7	14.4	4.4	20.7	0.3
Polar compounds (methanol)	30.8	24.4	78.3	5.0	34.4	0.4

[a] Results from HV particulate sampling.

[b] Mutagenicity was determined in Salmonella typhimurium TA98 with S9. Each sample was solvent exchanged to dimethyl sulfoxide and tested at a minimum of five doses in triplicate. The slope of the initial linear portion ($R^2 \geq 0.90$) of each dose-response curve was calculated by least squares linear regression. Data represent one home

that show higher mutagenicity and carcinogenicity than the wood or smokeless coal emissions. Wood combustion emits larger particles of lower organic content, including fewer and lower concentrations of PAH compounds. This study, consistent with the studies by Dekoning et al. (1984), suggests little association between domestic open-fire wood smoke and lung cancer. Smokeless coal, the other fuel used in the commune with a low rate of lung cancer (RS), showed low indoor concentrations of particles and a low organic content. In this study, the results of short-term bioassays for mutagenicity and carcinogenicity are in good agreement with the lung cancer mortality data. The multidisciplinary studies all point to an etiologic link between indoor smoky coal combustion, as opposed to smokeless coal or wood combustion, and lung cancer in Xuan Wei County.

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two fuels, especially smokeless coal, contained much lower percentages (Table 3). All indoor air samples contained more extractable organic matter than outdoor samples. Chemical analysis of the organic extracts of the smoky and smokeless coal samples showed that their major components were PAH and methylated PAH, whereas the major components of the wood samples were hydroxylated aromatic compounds, aliphatic aldehydes and alcohols, and PAH. Fewer PAH compounds were detected in the wood samples than in the coal samples. Quantitative analysis results (Table 4) revealed that indoor PAH levels were consistently higher in the smoky coal samples. The wood samples contained much lower levels of PAH, and the smokeless coal sample contained the lowest levels. Higher concentrations of methylated PAH were found in the smoky coal samples, which also contained higher levels of nitrogen heterocyclic compounds (Table 4). 1-Nitropyrene, a highly mutagenic nitro-PAH, was not detected in any of the samples. Filters in both the HV and MV samplers collected a wide range of PAH compounds, from three- to seven-ring compounds, whereas the XAD collected mostly compounds of four or fewer rings. The mass distribution results (Table 5) showed that extracts of indoor samples from coal combustion contained a higher percentage of aromatic compounds than the wood sample extracts; the smoky coal extracts contained the highest percentage. Most of the mass in the wood extract was in the polar fraction.

MUTAGENICITY AND CARCINOGENICITY

The smoky coal sample showed the highest mutagenic activity (Table 5). The wood sample displayed lower activity, and the smokeless coal sample displayed the least. The aromatic and polar fractions of the smoky coal samples contributed most of the mutagenic activity. The aliphatic fraction of all fuels was nonmutagenic. All XAD samples displayed little mutagenic activity.

In the mouse skin tumor initiation studies, the organic extract of the smoky coal sample induced more papillomas per mouse at the dose of 10 mg per mouse than the extracts of smokeless coal or wood samples (i.e., 7.5, 4.7, and 2.1 papillomas for smoky coal, smokeless coal, and wood samples, respectively) after 26 weeks of promotion with TPA. When applied dermally twice weekly for 52 weeks (total 2 mg per mouse per week) to the mice without TPA promotion, the smoky coal sample induced 0.50 carcinomas per mouse in the treated animals, but neither the wood samples nor the solvent-control samples induced any carcinomas at the end of 52 weeks.

SUMMARY

Physical, chemical, toxicological, and epidemiological studies all indicated that the emissions from smoky coal combustion are much more health-hazardous than the emissions from wood or smokeless coal combustion. Smoky coal combustion emits high concentrations of small (1-μm diameter) organic particles that contain PAH concentrations comparable to those typical of occupational exposure and

Table 4. Polycyclic aromatic hydrocarbon concentrations of indoor air particles (PM_{10}) during cooking in Xuan Wei homes.

Compounds	Concentrations (μg/m³)		
	Smoky Coal[a]	Wood[a]	Smokeless Coal[b]
Cyclopenta[cd] pyrene[c]	10.7 ± 1.5	5.6 ± 1.8	0.4
Benzo(a)anthracene[d]	25.1 ± 5.0	4.0 ± 1.1	1.0
5-Methylchrysene[d]	7.3 ± 2.7	0.2 ± 0.06	0.2
Benzofluoranthene[d]	22.7 ± 4.6	2.7 ± 0.7	1.9
Benzo(a)pyrene[d]	14.7 ± 3.0	3.1 ± 1.0	0.6
Indeno[1,2,3-cd] pyrene[d]	8.4 ± 3.9	2.0 ± 0.6	0.5
Dibenzo[ae]pyrene[d]	11.7 ± 4.2	0.2 ± 0.07	0.6
Dibenz-[a,j] acridene[b,d]	0.7	BDL[e]	0.007

[a] Data from four homes (mean ± SEM) by HV sampling.
[b] Data from one home by HV sampling.
[c] Compound showed limited evidence of carcinogenicity in experimental animals and sufficient evidence of mutagenicity/carcinogenicity in short-term bioassays (International Agency for Research on Cancer 1983).
[d] Compound showed sufficient evidence of carcinogenicity in experimental animals (International Agency for Research on Cancer 1983).
[e] BDL = below detection limit.